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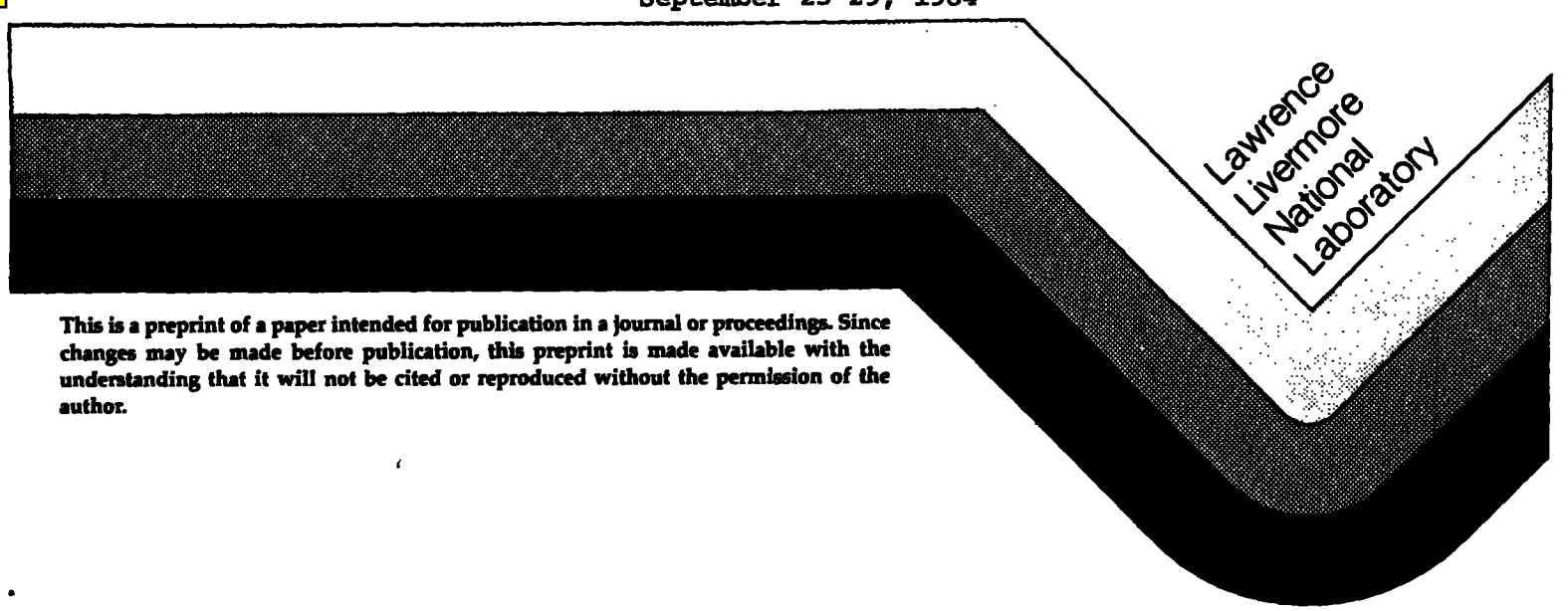

THE DEPOSITION OF SELENIUM COATINGS ON BERYLLIUM FOILS

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A technique for preparing selenium films on 50.8 micrometers thick beryllium foils is described. The selenium was deposited in vacuum from a resistance heated evaporation source. A water-cooled enclosure was used to minimize contamination of the vacuum system and to reduce the exposure of personnel to toxic and obnoxious materials. Profilometry measurements of the coatings indicated selenium thicknesses of 5.5, 12.9, 37.5, 49.8 and 74.5 micrometers. The control of deposition rate and of coating thickness was facilitated using a commercially available closed-loop programmable deposition controller. The x-ray transmission of the coated substrates was measured using a tritiated zirconium source. The transmissivities of the film/substrate combination are presented for the range of energies from 4 to 20 keV.

1. Introduction

The interest and demand for both free-standing and coated foils for use in nuclear physics experiments have encouraged the development of numerous preparation techniques. A variety of methods have been used to deposit pure or alloyed films of selenium: Jansen [1] developed a novel

apparatus fashioned after the flash evaporation technique which allows the positive control of the deposition rate of low melting point (As/Sb/Se) alloys; Johnson and Schlie [2] deposited thin films of zinc, selenium, and nonstoichiometric ZnSe by photodissociation with UV energies; even electrochemical deposition methods [3] have been used. Physical vapor-deposition methods such as reactive sputtering [4] and evaporation from electron beam [5] and resistance heated [6-8] sources have also been applied with good results.

Because of its inherent simplicity, the resistance heated source was selected to produce selenium coatings on beryllium substrates. Early test runs had indicated that, although the quality of the coatings was adequate, the integration of closed-loop deposition control would be advantageous. Since selenium has a relatively high vapor pressure (13 Pa @ 277° C) [9], and presents a potential hazard to humans [10], it is desirable to control the amount of material introduced to the vacuum system. A enclosure was designed to reduce the contamination of the vacuum system.

2. Equipment and Procedure

The evaporator used to deposit selenium coatings consisted of a typical LN_2 trapped, oil diffusion pumped, vacuum system (fig. 1). The walls of the stainless steel chamber were protected from the deposition of selenium by the use of a water-cooled enclosure. The evaporation source, the substrates, a shutter, and the deposition monitor were located within the enclosure. The enclosure was constructed of 304 stainless steel with water cooling passages welded to the outer surfaces. The interior dimensions of the enclosure were 30 cm wide by 30 cm deep by 50 cm high.

A resistance heated tantalum source was used to evaporate the selenium. The source, of an open-boat design, had sufficient capacity for the deposition of 50 micrometers thick films when fully charged with 100 grams of selenium shot [11]. The source temperature was monitored with an intrinsic chromel-alumel thermocouple, spot-welded to the bottom of the boat. A programmable deposition controller [12] was used to control the source power supply in a closed-loop control of preheat time and temperature, deposition rate, and deposited thickness.

The substrates, in this case beryllium sheet material approximately 3 cm diameter and 50.8 micrometers thick, were attached to an aluminum holder. The holder was in turn clamped to the water-cooled lid of the containment enclosure. The source to substrate distance was about 37 cm, and the substrates were arranged within a 15 cm diameter circle. Although the temperature of the substrates was not monitored during deposition, the estimated maximum temperature was about 40° C. No substrate motion was used during the deposition. However, because many of the coatings were too thick to be deposited in a single coating run, the coating unformity was improved by manually rotating the substrates while the system was at atmosphere and the evaporation source was being reloaded. The amount of rotation depended upon the number of coating runs which were required (i.e. 180 degrees for 2 runs, 120 degrees for 3 runs, etc.).

A shutter was used to protect the substrates and the quartz crystal monitor from contamination during the initial heating and degassing of the source material. The shutter was actuated by the deposition controller, which opened the shutter after a timed preheat and closed the shutter

after the desired deposit thickness was reached. The measured temperature of the evaporation source during a typical coating run is shown in fig. 2.

The coating thickness deposited during each of the runs was determined using stylus profilometry [13]. Polished glass slides were included in every coating run and were used as witness pieces. The thickness of the coating deposited on the glass slide was representative of the coatings deposited on the beryllium substrates.

Two other techniques were used to verify the total thickness of the selenium coatings: gravimetric and computation of x-ray density. The first technique, the measurement of the weight gain of a given substrate due to the deposition of a coating over a known and calculable area, depends upon the assumption of bulk density in order to estimate the coating thickness. The second technique also requires the assumption of certain bulk properties such as density and x-ray cross section. The x-ray density technique is a by-product of the x-ray measurements normally performed on each of the coated foils.

3. Results and Discussion

The use of the controlled deposition rate technique proved to be a major improvement over previously used methods. Prior to integrating the closed-loop control system, the deposition method depended upon the complete evaporation of preweighed charges of evaporant. In addition to the lack of real-time monitoring of coating thickness, the major drawback of this method was the inability to properly degas the evaporant prior to the start of the deposition cycle. In applying the closed-loop control

system, preweighed charges were no longer required. The melting and degassing could proceed as necessary, and the coating process could be terminated upon reaching a desired (and measurable) thickness. The nominal deposition rate used for these coatings was on the order of 20 to 21 nm per second to a maximum thickness of about 35 micrometers during any one run.

The use of the water-cooled enclosure as a means of reducing the contamination of the vacuum system was also successful. During nearly 20 coating runs, many of which required the deposition of relatively thick selenium coatings, the chamber walls remained clean, and the vacuum pumping system was not affected by the deposited material. Pump down times and base pressures were substantially the same regardless of the thickness of the coatings which were deposited on the inner walls of the enclosure. Deposited selenium was localized and contained within the enclosure, which minimized the exposure of personnel to a toxic and obnoxious material and reduced maintenance requirements.

The manual rotation of the substrates between coating runs and the favorable source-to-substrate distance apparently combined to good advantage. The coating uniformity, measured by a nonquantitative beta-backscattering method [14] was within 5% of the desired coating thickness.

Comparing the results of stylus profilometry and gravimetric measurements indicated that the density of the coatings was about 98% of the bulk density of vitreous selenium. However, the computation of the coating density by means of the x-ray transmission and photo-interaction

cross sections indicated that the coating densities were more on the order of 90% of bulk. While it is not entirely clear at the present time just which measurement is most correct, the vacuum deposition of thin films can often result in coatings which are of less than bulk density. While there is interest in resolving the disparity in the measurements, in our present application of these coatings the most important data is the transmission of x-ray energies through the selenium-beryllium (coating-substrate) combination.

The x-ray transmission of the coated substrates was measured over the range of 3 keV to about 15 keV using a tritiated zirconium source [15,16]. Since the beryllium substrates are essentially transparent to x-rays over this energy range, the transmission values are that of the selenium coatings. Each of the coated foils was calibrated thusly, which provided the experimenter with the particular characteristics of every coating. The percentage x-ray transmission as a function of incident energy for five different thicknesses of selenium coatings is shown in fig. 3.

4. Summary

The integration of a simple, "tried and true", method of deposition with a modern, closed-loop thin film control system has been illustrated. The use of a water-cooled enclosure during the deposition of a high vapor pressure material such as selenium has proved effective in minimizing deleterious effects upon the vacuum coating system. The use of

the enclosure also reduced maintenance and system cleanup requirements. The same scheme might be expanded to include other applications. The thick selenium films produced under these conditions were quite uniform, but some questions remain unanswered regarding the actual coating density. The x-ray transmission of selenium coated beryllium foils was measured over the range of energies from 3 keV to 15 keV.

5. Acknowledgements

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13. Sloan Dektak IIA, Sloan Technology Corporation, Santa Barbara, California 93103.

14. TCI Model TC-3000 Betascope, Twin City International, Incorporated, Amherst, New York 14150.

15. For further information concerning LLNL's x-ray measurement capabilities, contact:

    X-ray Calibration and Standards Laboratory
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    Livermore, California 94550, U. S. A.

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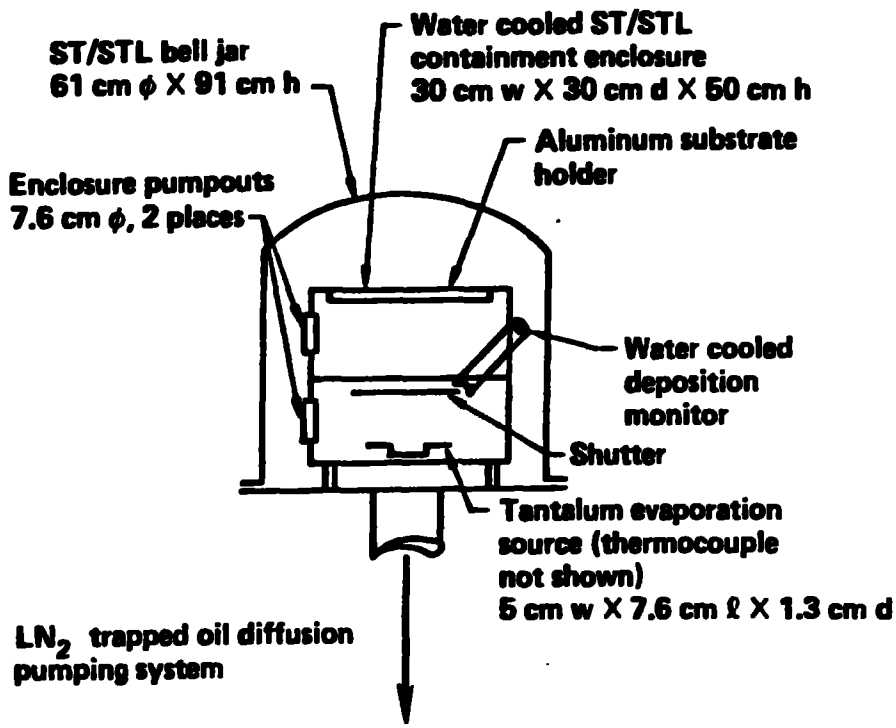

Fig. 1 A schematic diagram of the evaporation system and the containment enclosure which was used to deposit selenium coatings.

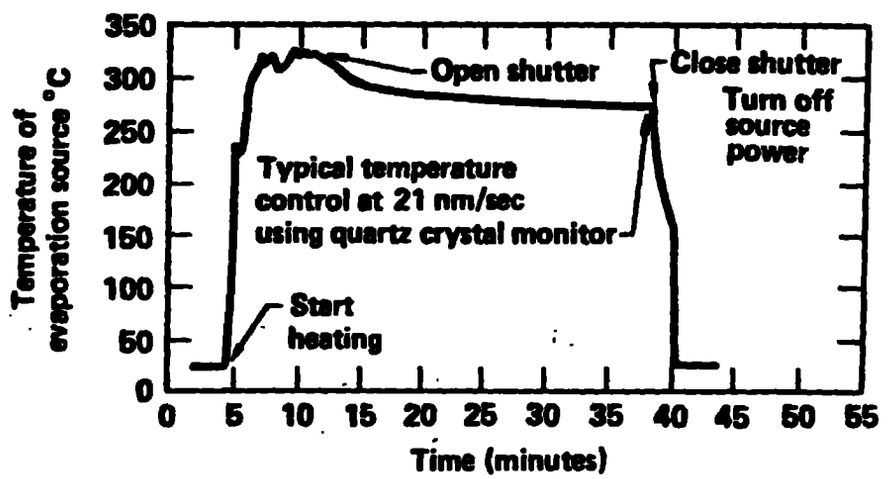

Fig. 2 A graph of the temperature of the evaporation source as a function of time during a typical closed-loop coating cycle at a controlled deposition rate of 20-21 nm per second.

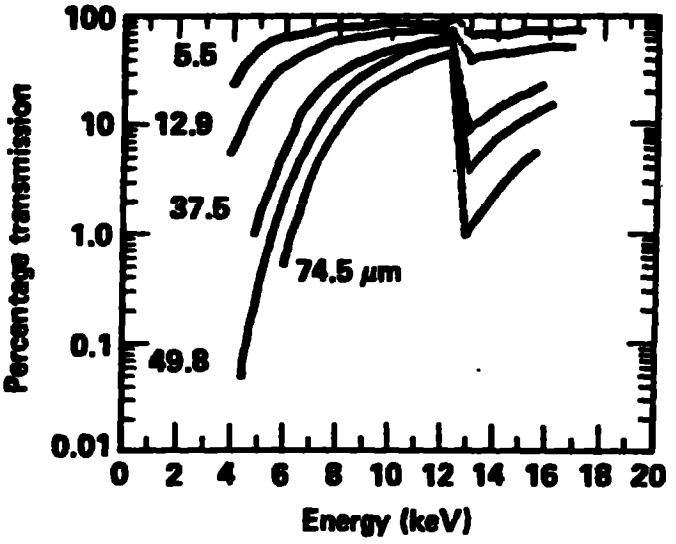

Fig. 3 A graph of the measured x-ray transmission of selenium coated beryllium foils as a function of incident energy. The substrates were approximately 50.8 micrometers thick. The thickness of the selenium coatings is as indicated.